Inner-Shell Photodetachment of Negative Ions
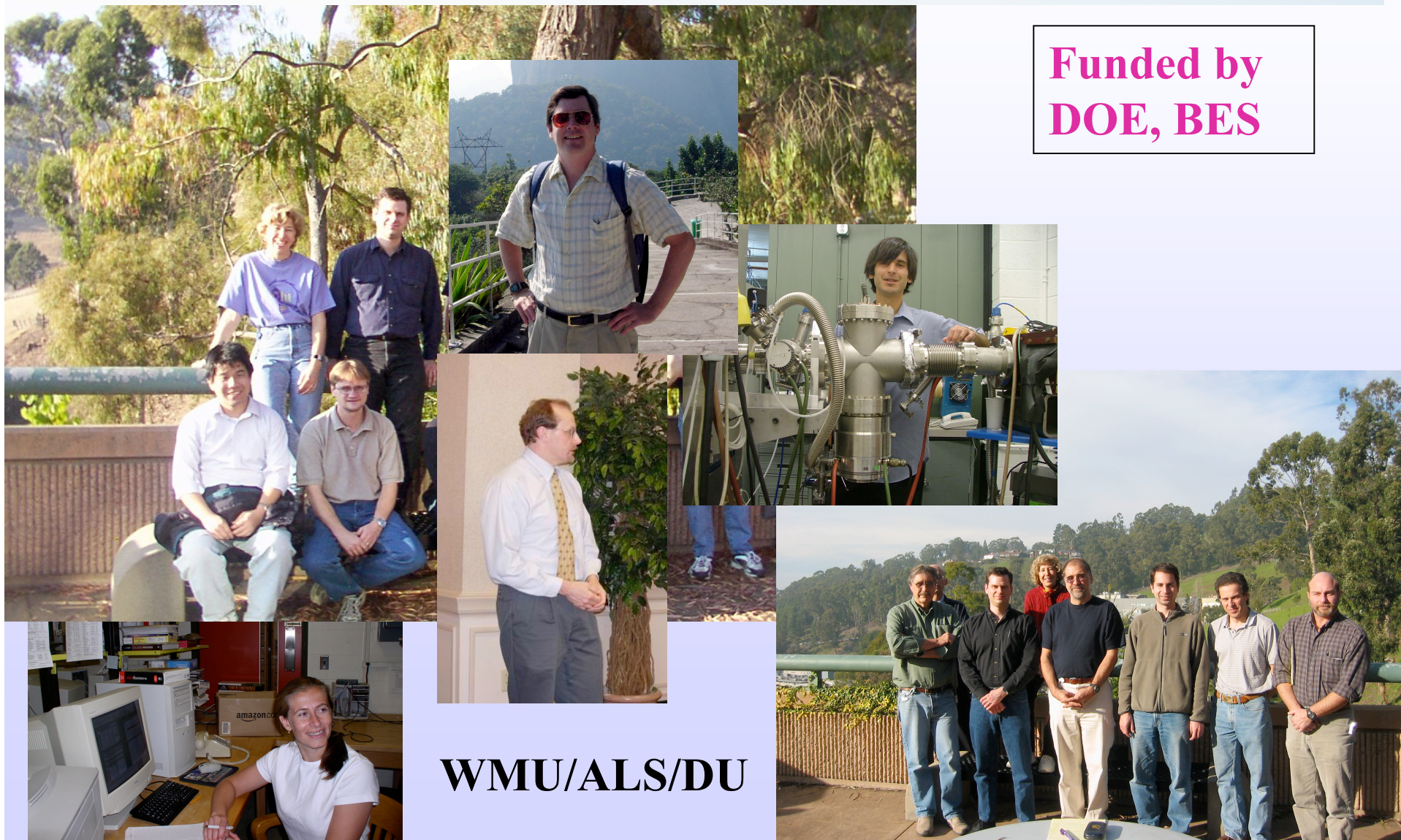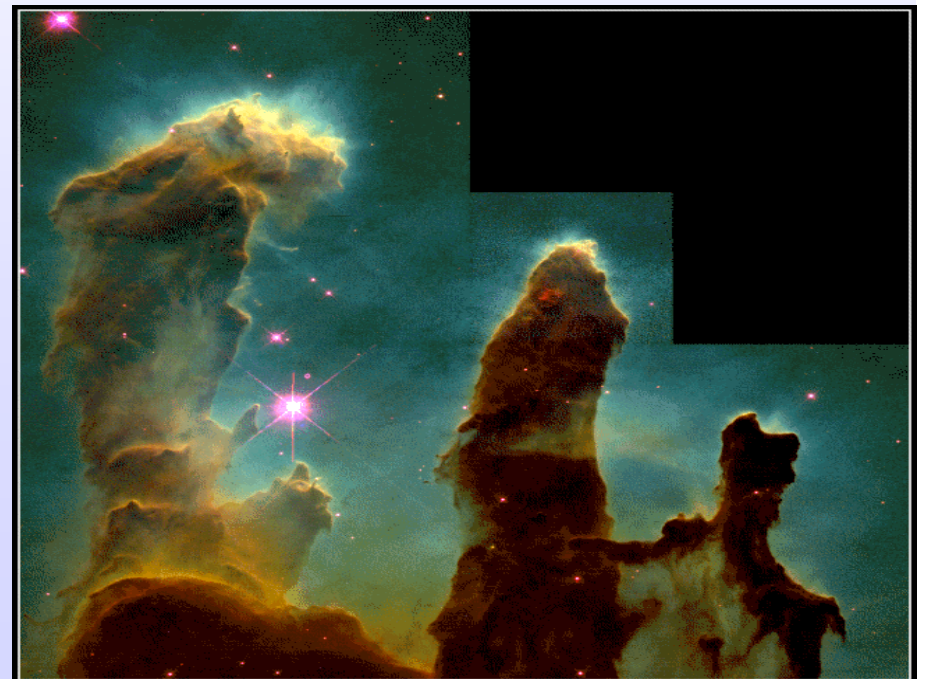
WMU/ALS/DU

<u>Outline</u>

- **General Introduction**

- **Simultaneous Double Auger Decay, He$^-$**

- **Threshold Phenomena**

- **Production of Highly Charged Ions, S$^-$**

- **Photodissociation of small clusters**

Why Study Negative Ions

•Offer New Perspective for Understanding Strongly Correlated System.

• Theoretical and Experimental Challenge.
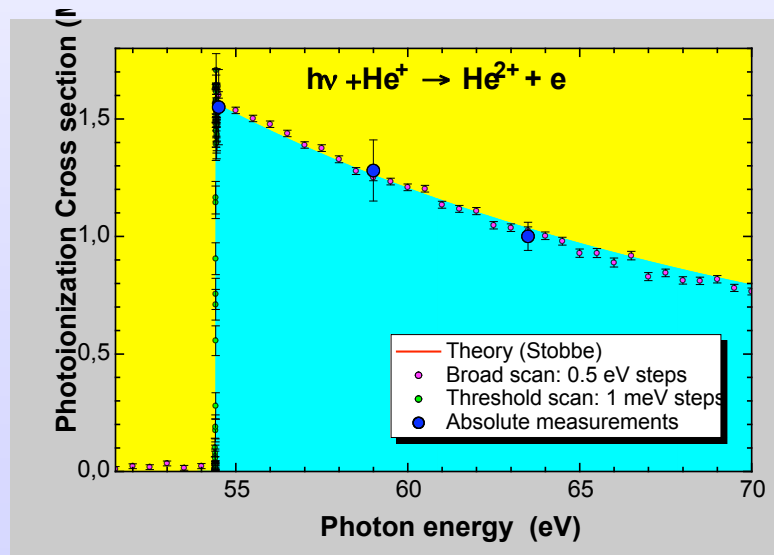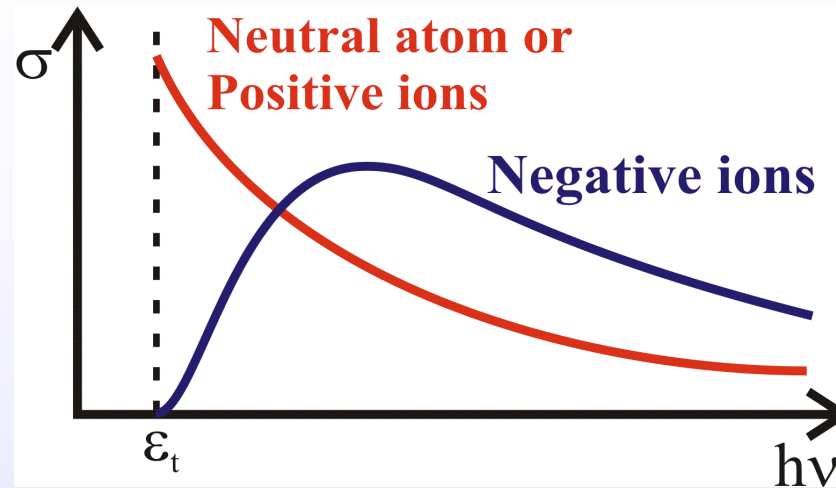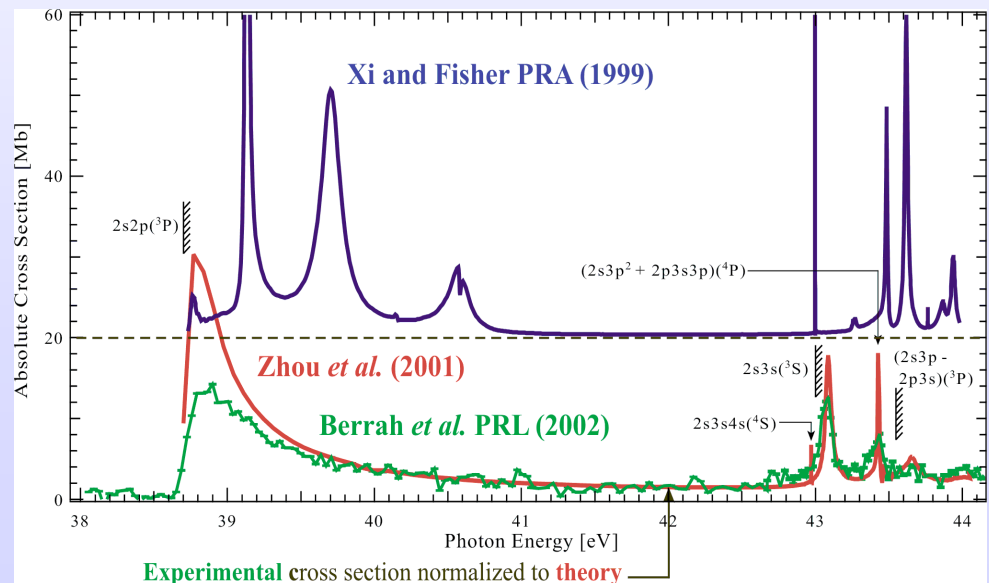

Gaseous Pillars · M16   HST · WFPC2
PRC95-44a · ST ScI OPO · November 2, 1995
J. Hester and P. Scowen (AZ State Univ.), NASA

How different are the Photodetachment of Negative Ions and Photoionization of Neutral Atoms/ Positive Ions???
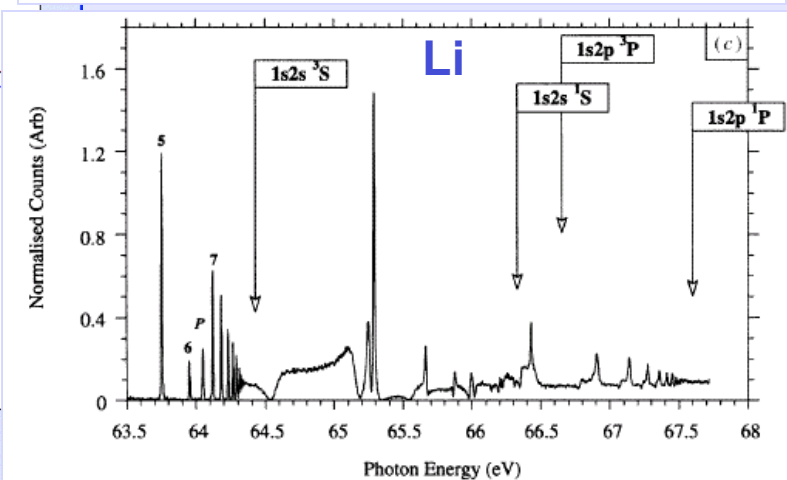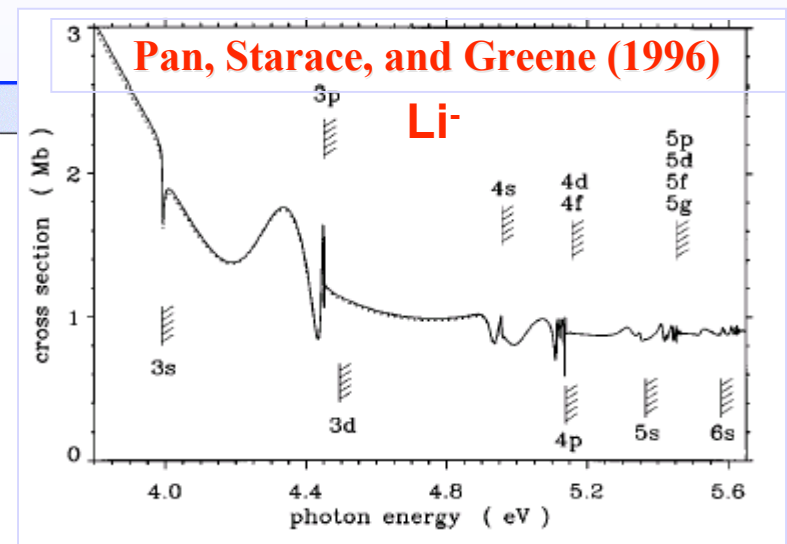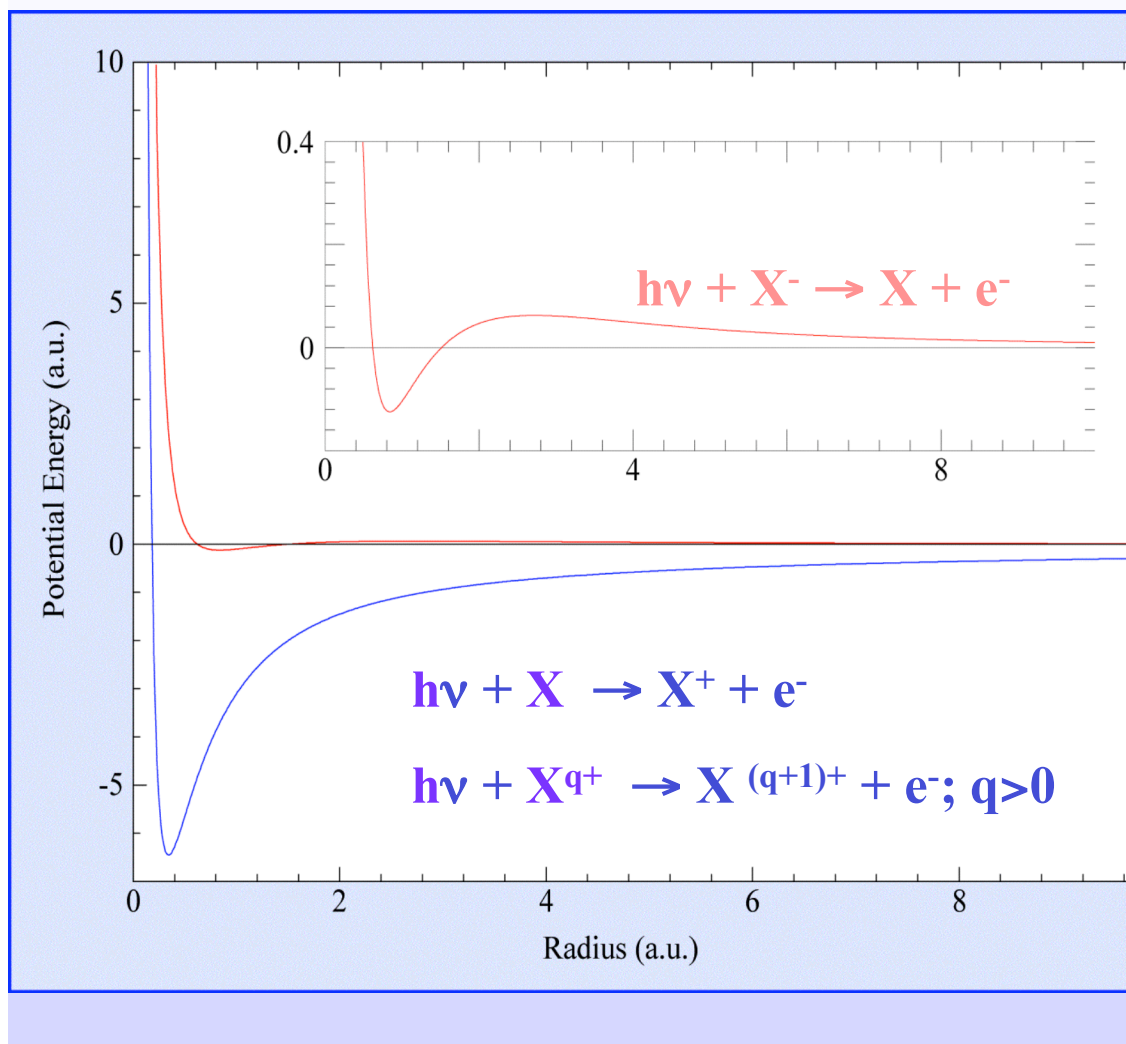

Photoionization of He

Photodetachment of He⁻

Coulomb interaction is effectively screened in negative ions ⇒ Different dynamic, structure and properties….

WHY?

$$h\nu + X^- \rightarrow X + e^-$$

$$h\nu + X \rightarrow X^+ + e^-$$

$$h\nu + X^{q+} \rightarrow X^{(q+1)+} + e^-; \; q>0$$

Pan, Starace, and Greene (1996)

Li⁻

Kiernan et al. (1996)

Experimental Apparatus of the ...amline @

Size Selected Production

Size and Charge Selected Detection

Beam merger (electrostatic)

Steering, focussing

Focussing, BPM's & interaction region

I^+ detector

m/q selector (magnetic)

Negative Ion Sources
- SNICS-II
- Alphatross

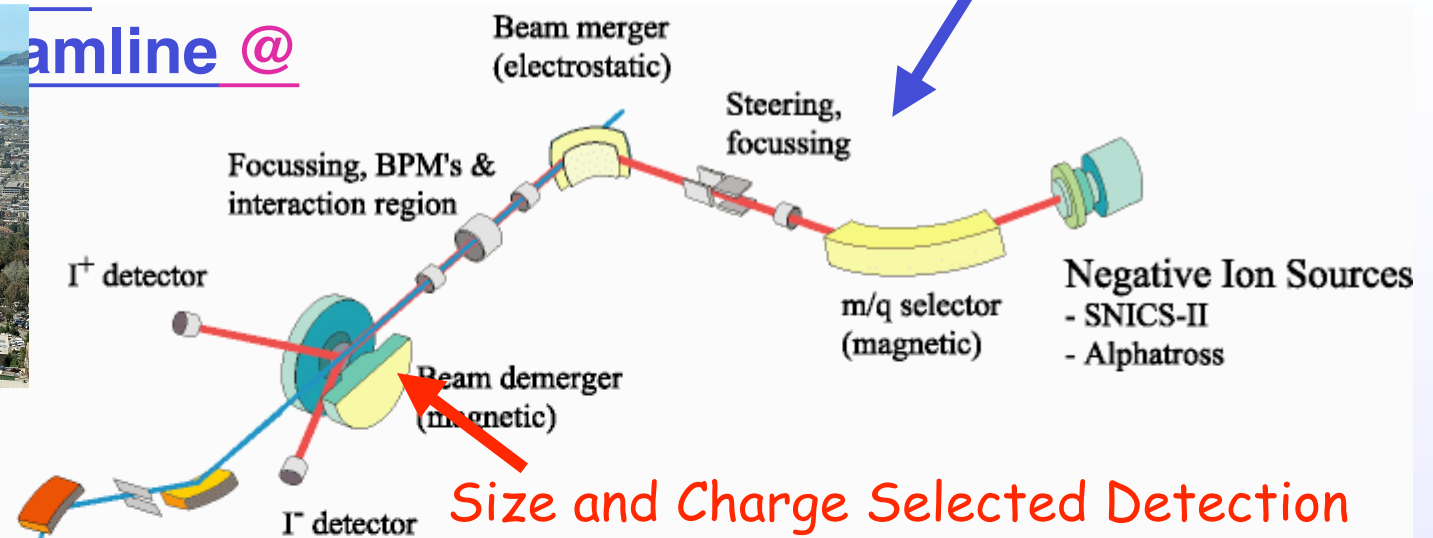Beam demerger (magnetic)

I^- detector

Beamline 10.0.0
- SGM
- high resolution (>10,000)
- high flux (>10^{12} ph/s)

ALS
High brightness undulator source

- Photon beam
- Ion beam

x   y
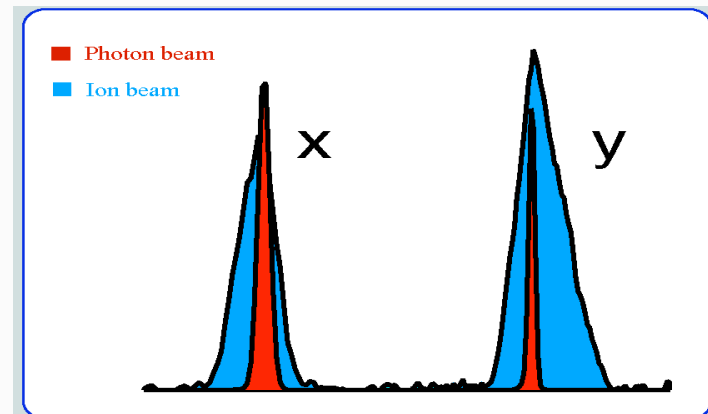
Absolute cross sections of ions available from careful measurements of overlaps, photon & ion fluxes and

K-Shell Photodetachment of

$He^- 1s2s2p\ ^4P°$

$h\nu + He^- \rightarrow He + e^-$

$\rightarrow He^* + e^-$

$\rightarrow He^+ + e^-_A + e^-$

Probing Doubly and Triply Excited States!!

Hollow Ion/Atom

K-Shell Photodetachment of He⁻
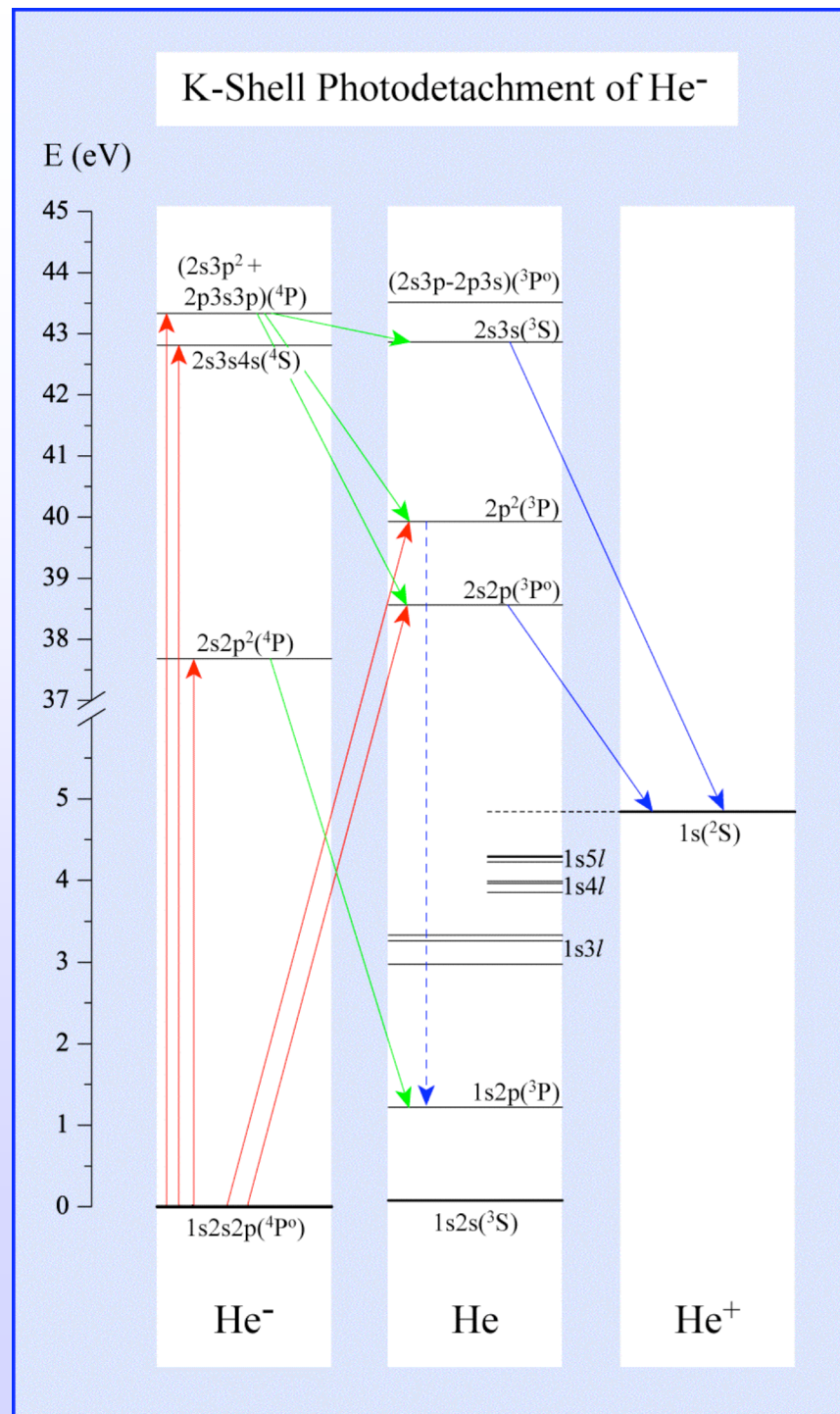
K-Shell Photodetachment of He⁻ :Comparison Between Two Calculations in Dispute + PCI Effect

Xi and Fisher PRA (1999)

$2s2p(^3P)$

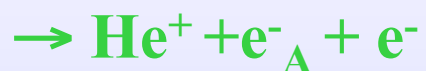$(2s3p^2 + 2p3s3p)(^4P)$

PCI

Zhou *et al.* (2001)

$2s3s(^3S)$

$(2s3p - 2p3s)(^3P)$

Berrah *et al.* PRL (2002)

$2s3s4s(^4S)$

Absolute Cross Section [Mb]

Photon Energy [eV]

Experimental **c**ross section normalized to **theory**
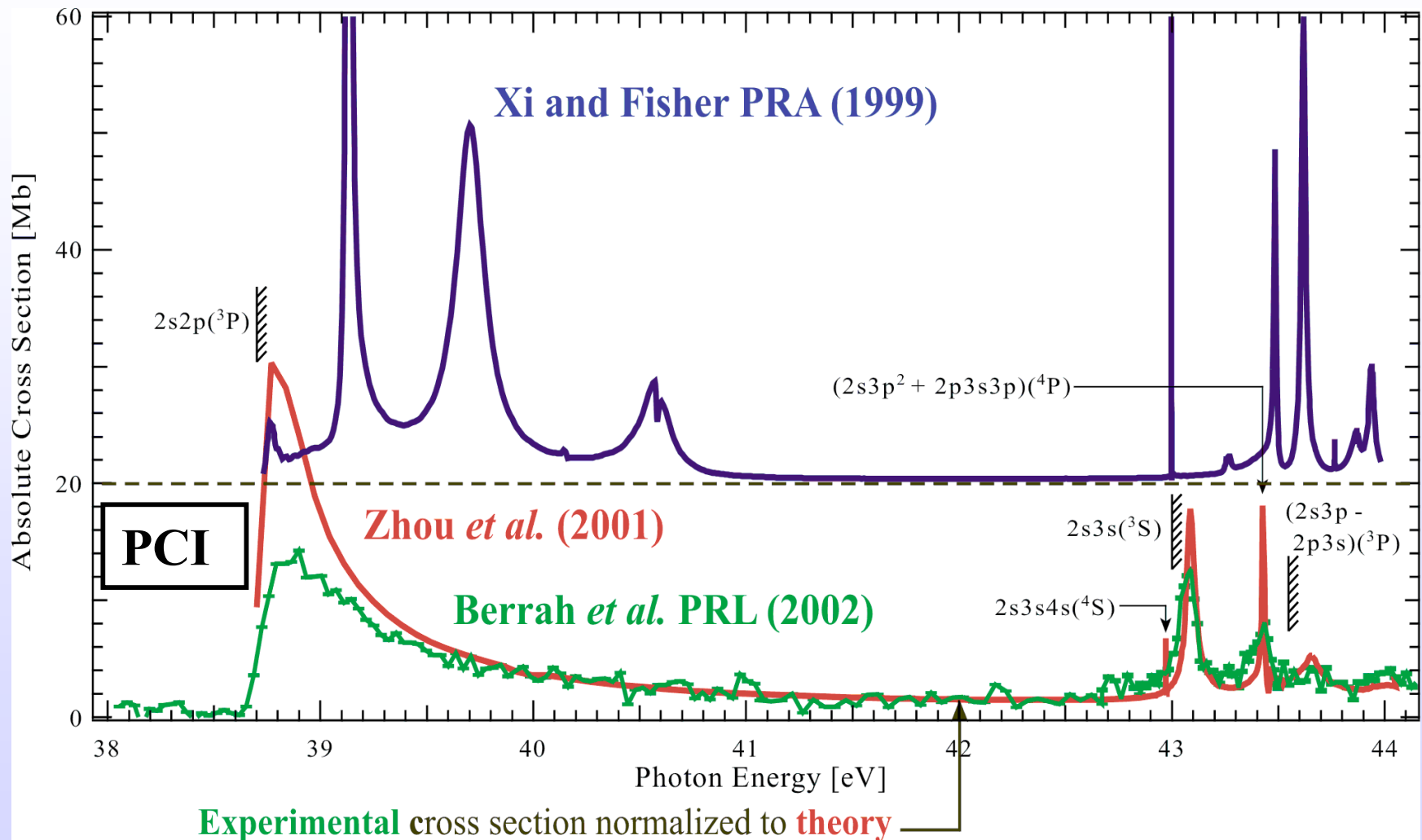
Berrah, Bozek, Turri, Ackerman, Rude, Zhou, Manson, PRL 88, 093001 (2002).

Search for the 2s2p² ⁴P State of He⁻ ???

$h\nu + He^- (2s2p^2\ ^4P) \rightarrow$??
$He^+\ (1s\ ^2S) + e^- + e^-$
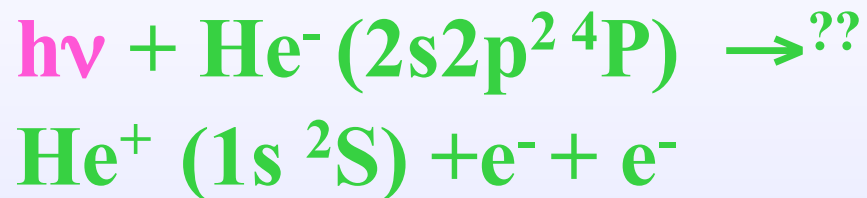
Probe Feshbach Resonance via
Simultaneous 3e⁻ Decay Process!!?

K-Shell Photodetachment of He⁻

E (eV)

$(2s3p^2 + 2p3s3p)(^4P)$
$(2s3p-2p3s)(^3P^o)$
$2s3s(^3S)$
$2s3s4s(^4S)$
$2p^2(^3P)$
$2s2p(^3P^o)$
$2s2p^2(^4P)$
1%
$1s(^2S)$
$1s5l$
$1s4l$
$1s3l$
$1s2p(^3P)$
$1s2s2p(^4P^o)$
$1s2s(^3S)$
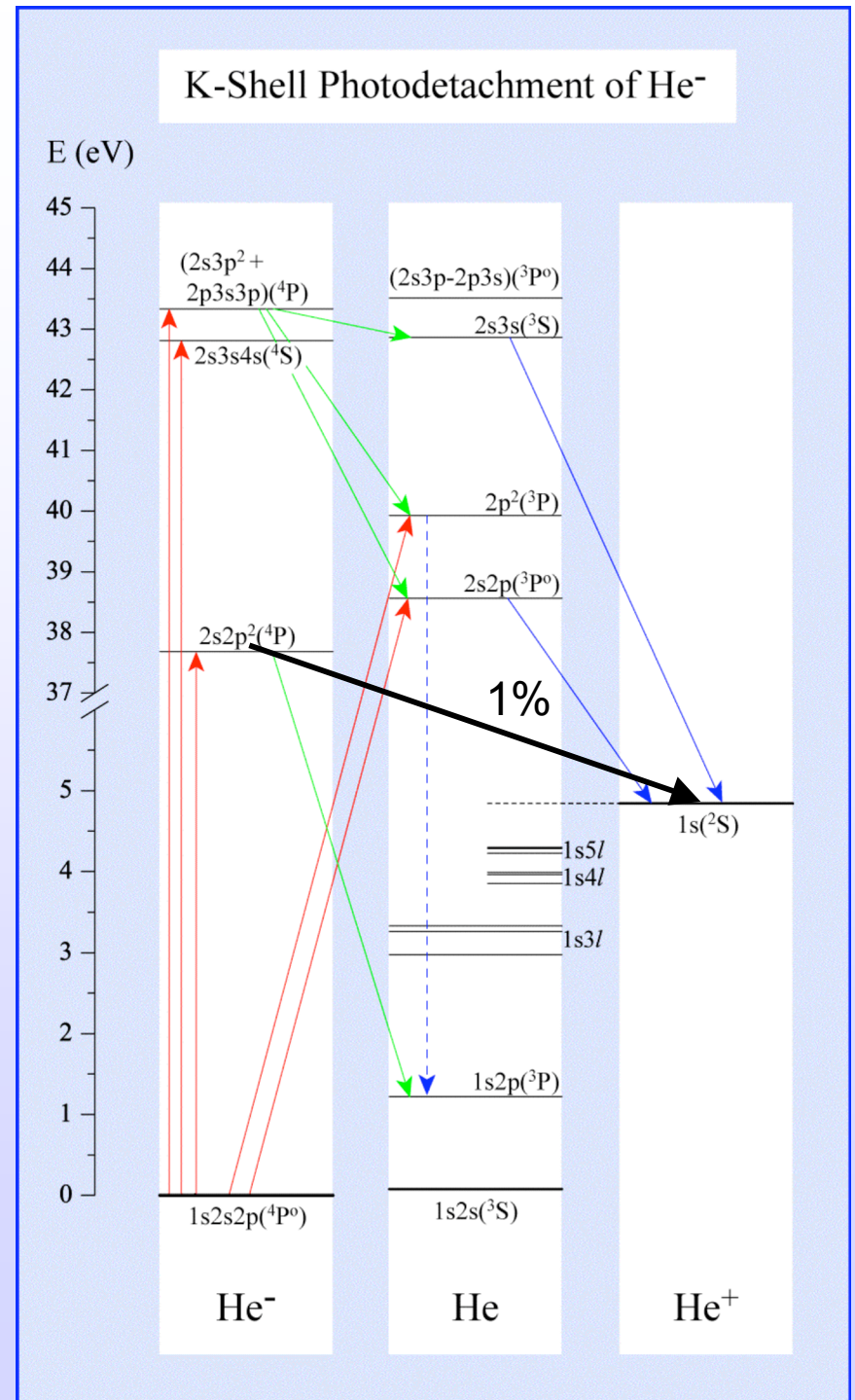
He⁻   He   He⁺

Feshbach Resonance: Observation of the $2s2p^2$ ^4P State of He$^-$

Strong Double Auger Decay!! 11%
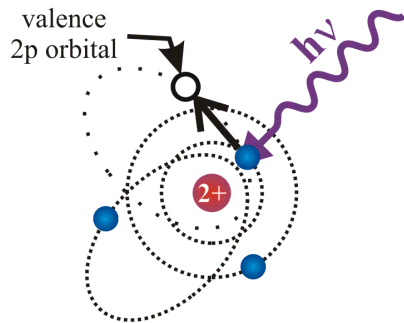
He$^-$ 1s Excitation
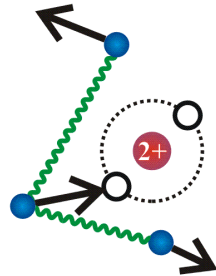
Double Auger Decay
(3-electron interaction)

First Evidence of Simultaneous 3 e$^-$ Decay process in Core-Excited Negative Ions

He$^+$ Signal

$2s2p^2$ ^4P

$\_$ =10 meV
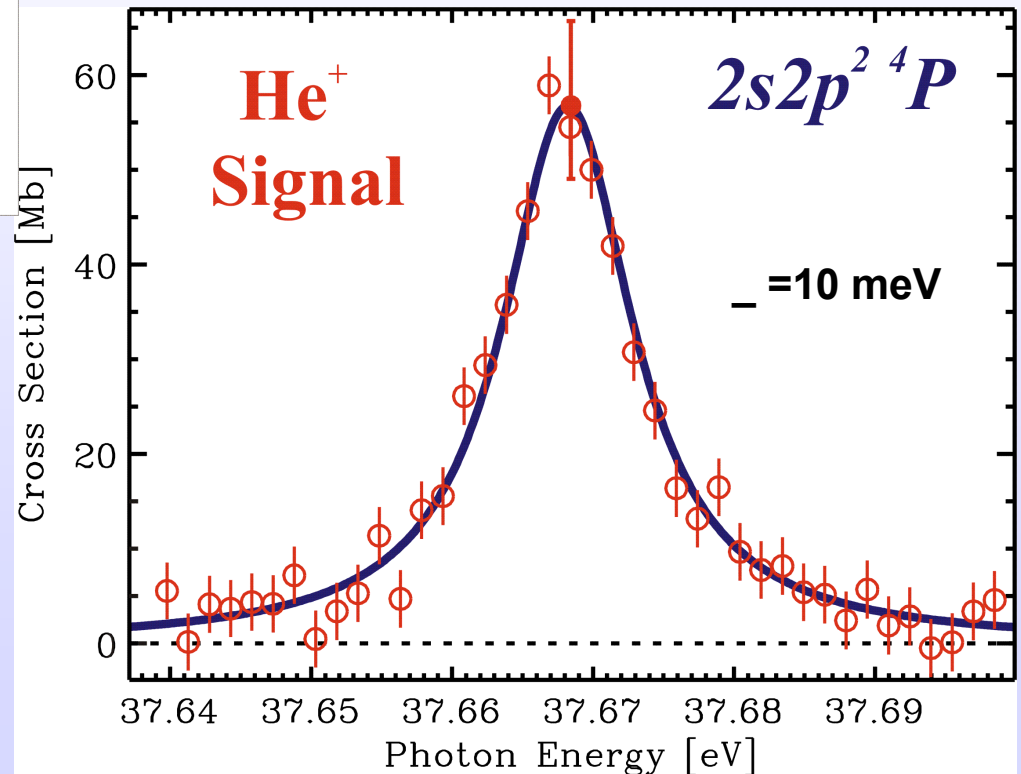
Bilodeau, Bozek, Turri, Ackerman and Berrah PRL, 93, 193001 (2004)

Threshold Law: TheWigner Law

$$\sigma = \sigma_0 \varepsilon_e{}^{l + 1/2}$$

$\sigma_0 =$ amplitude

$\varepsilon_e = h\nu - \varepsilon_t$;photoelectron energy

$\varepsilon_t =$ threshold energy

$l = l_0 \pm 1$; l_0 angular momentum of the bound electron being detached

s-detachment, $l_0 = 0$, $l = 1 \Rightarrow$ p outgoing wave (He⁻)

p-detachment, $l_0 = 1$, $l = 1 \pm 1$, s , d outgoing wave (S⁻)

Test Validity of Wigner Law in Inner-Shell Photodetachment of Negative Ions???

$$\_ \sim (h\_ - \_t)^{l+1/2}$$

Electron Angular Momentum

s-wave

p-wave

Threshold Energy

Negative ions

Neutral atoms and positive ions

Cross Section

Photon Energy

Wigner *p*-wave law fit He⁻ ??

$$\sigma \propto (h\nu - \varepsilon_t)^{3/2}$$

1s photodetachment in
He⁻ (1s2s2p ⁴P)
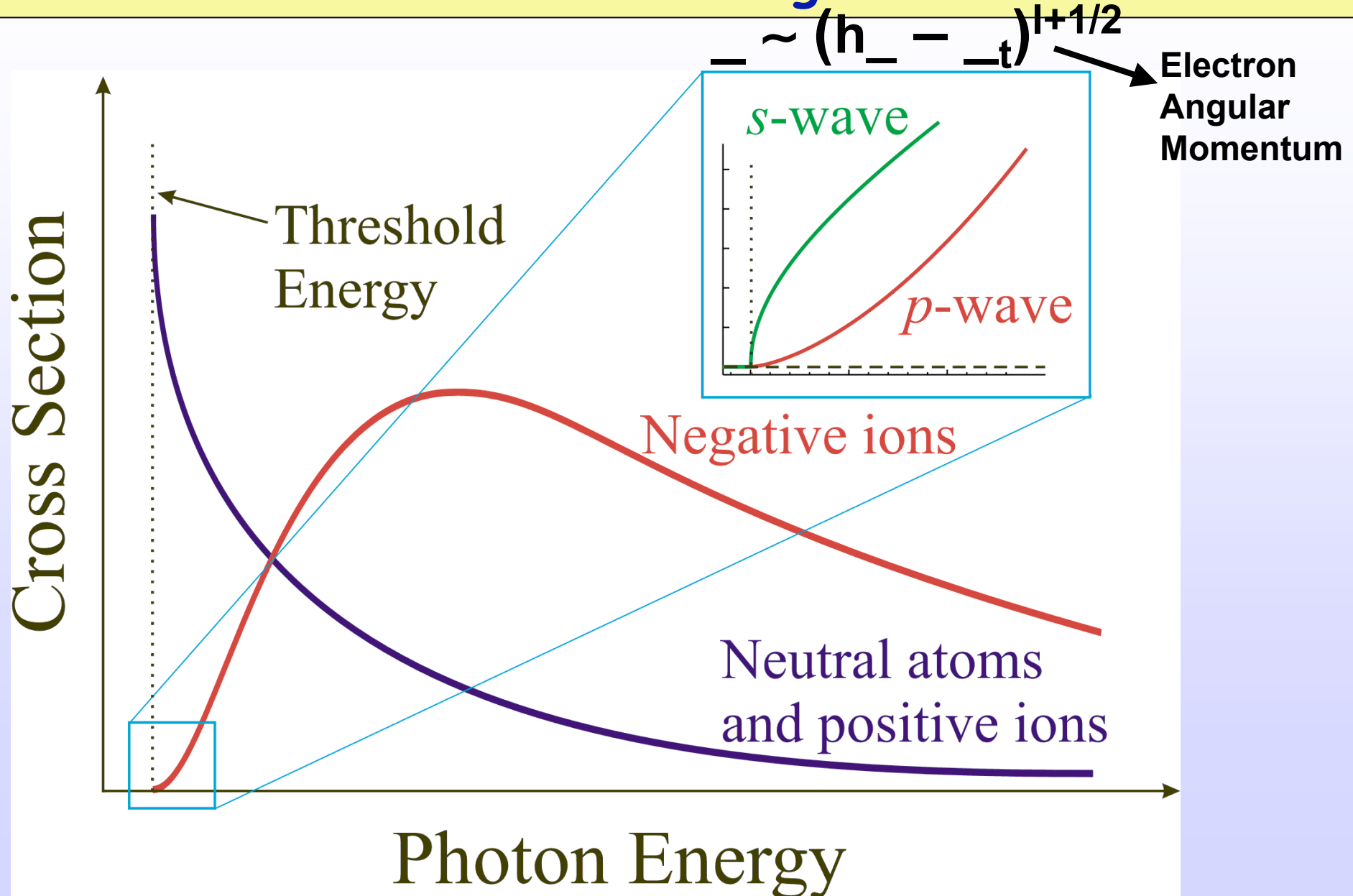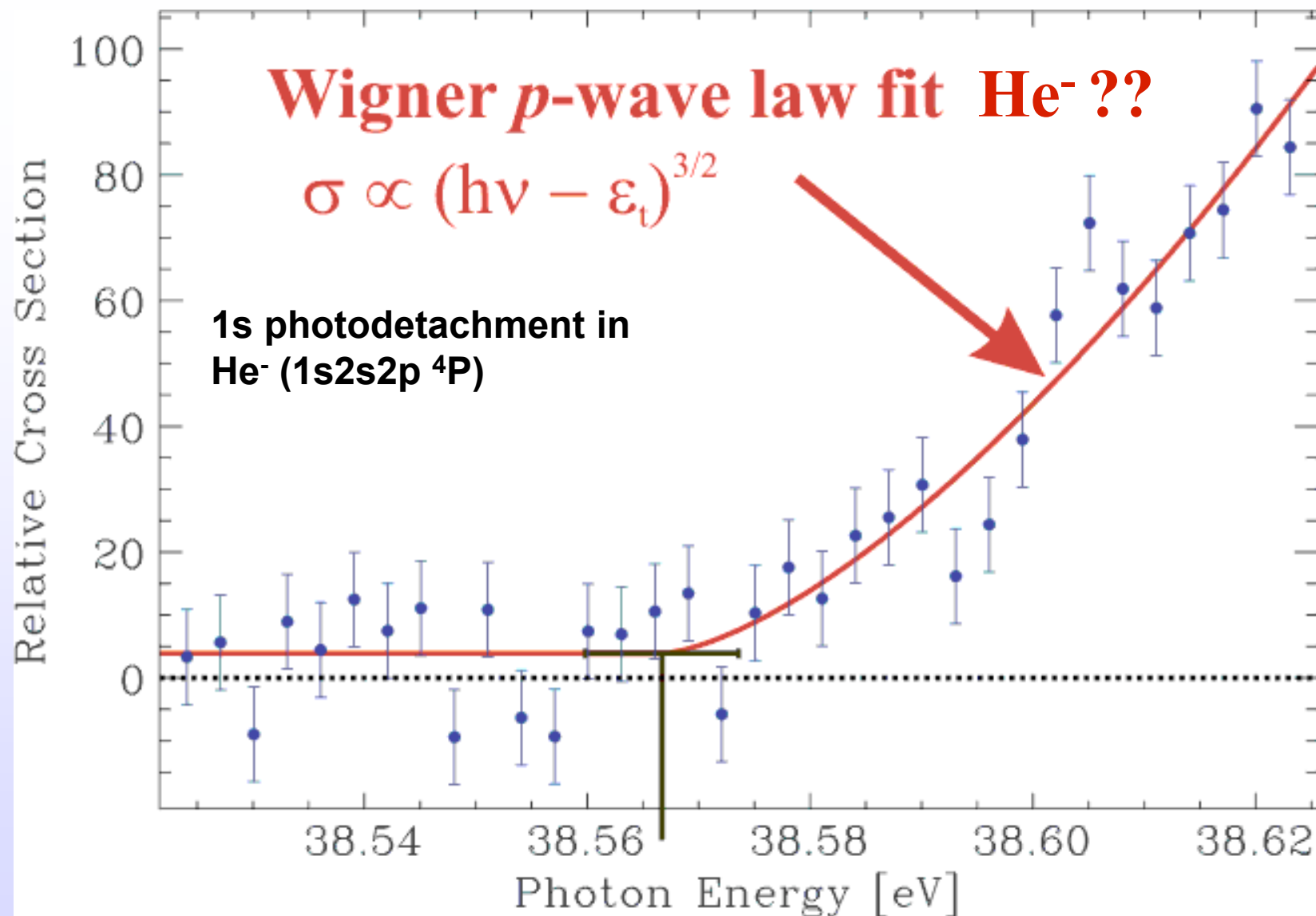
Why doesn't PCI affect the validity of the Wigner law??

K-shell Photodetachment Threshold of 1s2s3p ^{4}P in He$^-$

-.-.Vicario,no PCI   …Vicario, PCI — p-Wave Wigner Law Fit

2s2p ^{3}P^0

PCI ⟹Shifts σ

σ Shape not Altered by PCI

Predicted Threshold Energy
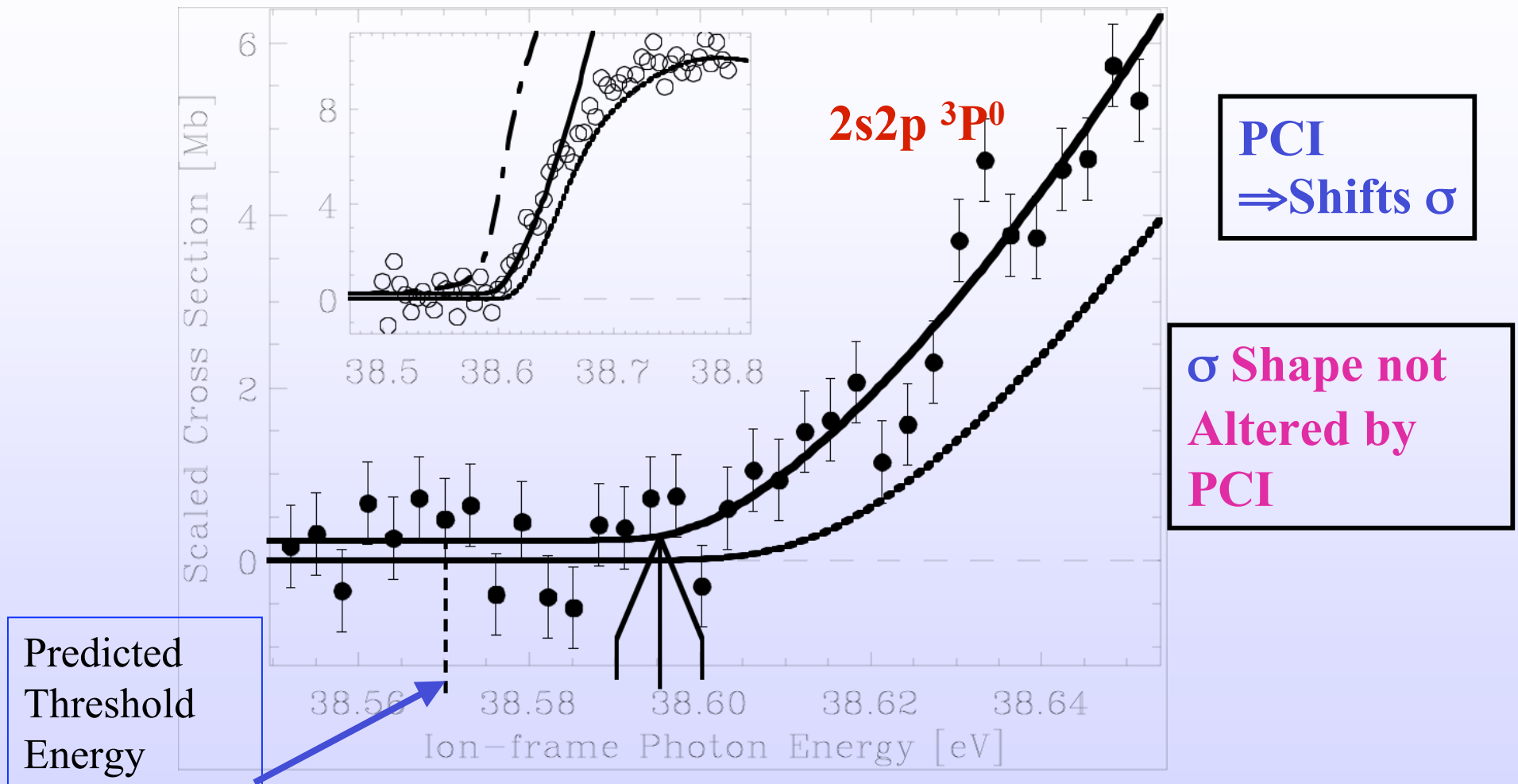
Bilodeau, Bozek,Gibson,Walter, Ackerman, Dumitriu, and Berrah, Phys. Rev. Lett. 95, 083001 (2005).

What Type of Phenomena or Processes Occur in the case of Inner-Shell Photodetachment of Heavier Targets???

High Charge State Formation Following 2p Photodetachment of S⁻

Title and figure labels:

High Charge State Formation Following 2p Photodetachment of S⁻

Figure axes:
- y-axis: Cross Section [Mb]
- x-axis: Ion-frame Photon Energy [eV]

Data series labels: S^+, S^{2+}, S^{3+} (3×), S^{4+} (50×)
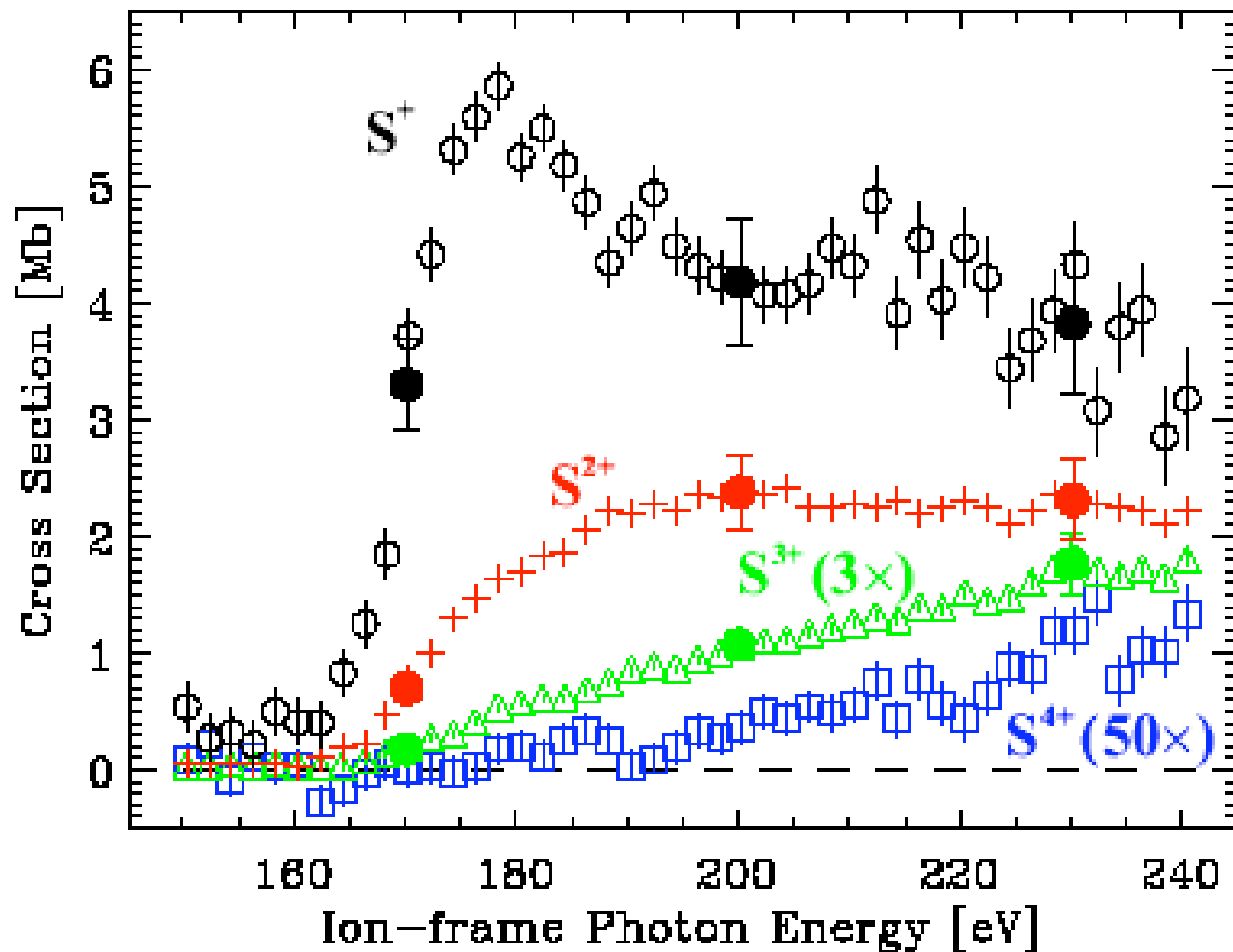
Side boxes:
- $S^+/S^{2+} \sim 60\%$
- $Li^{3+}/Li^{2+} < 1\%$
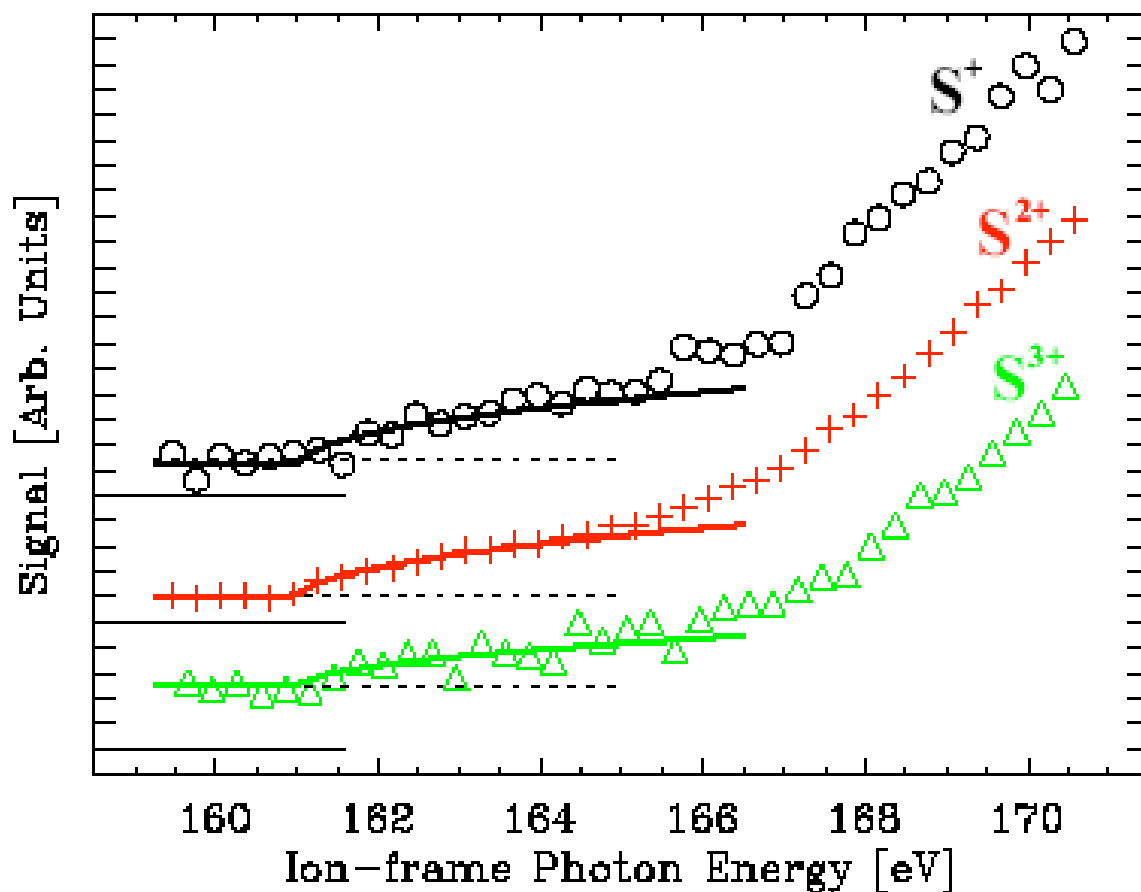- Th, Sim-Auger
- Int, K-Out
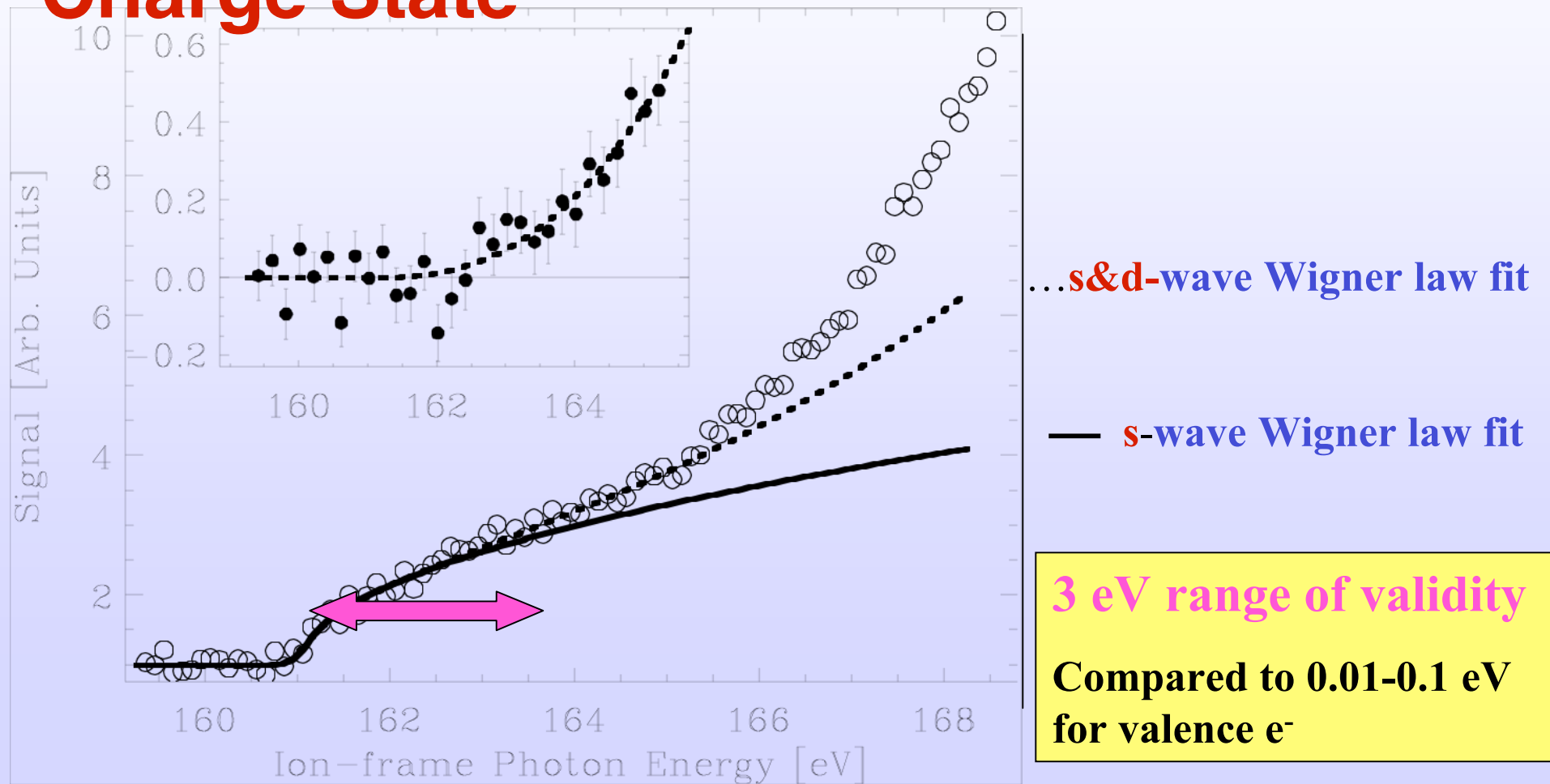- H, S-Off; or S-Up+Seq-Aug

2p Photodetachment Threshold for S^+, S^{2+} and S^{3+}

s-Wave Wigner Law Valid for All Channels

2p Threshold Photodetachment in S⁻

Threshold investigation in S^{2+} Charge State

...**s&d**-wave Wigner law fit

— **s**-wave Wigner law fit

3 eV range of validity

Compared to 0.01-0.1 eV for valence e⁻

2p Threshold Photodetachment in S⁻.

Threshold investigation in S⁺ and S³⁺ Charge States
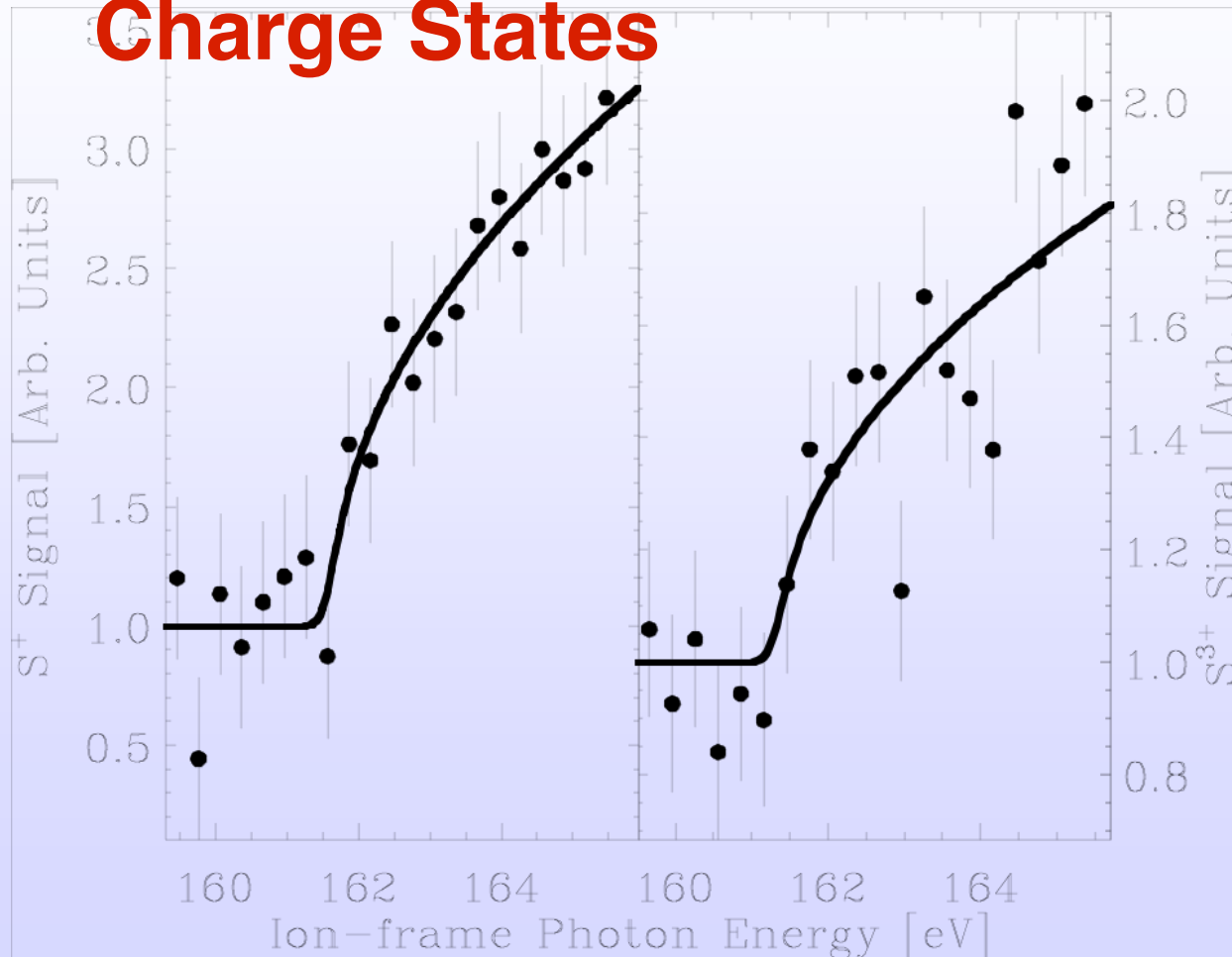
— s-wave Wigner law fit

Bilodeau, Bozek, Ackerman, Gibson, Walter, Aguilar, Turri, Dumitriu and Berrah PRA rapid Comm (in press)

2s Photodetachment Threshold for S^+, S^{2+} and S^{3+}

2s_3p Feshbach resonance, below 2s threshold in all three Channels!?

Fano profile +

p-wave Wigner law
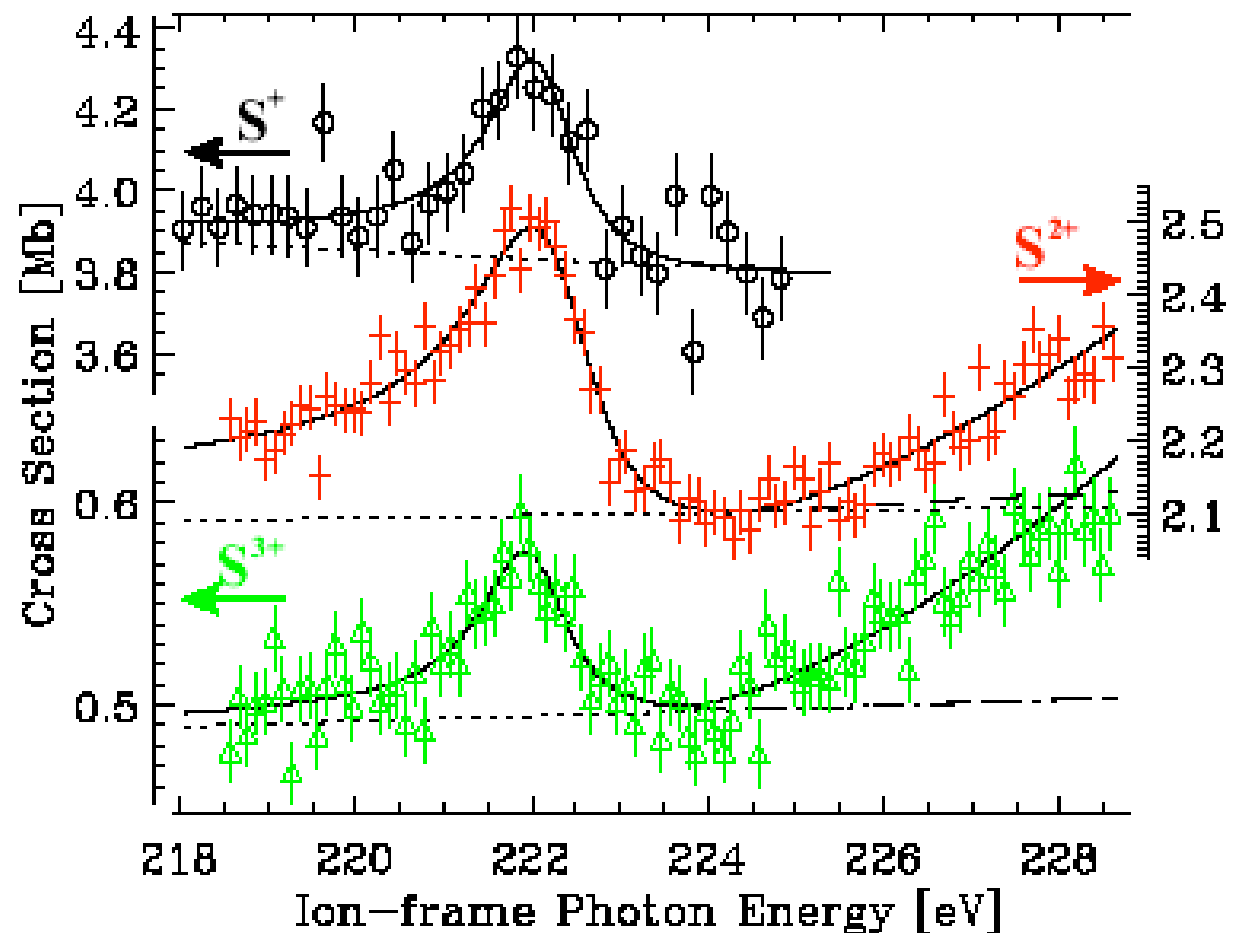
1s Ionization of size selected B_n^-

B^- $1s^2 2s^2 2p^2$ ^3P

Competition:
Photodetachment &
Photodissociation
in B_2^- and B_3^-

Sum of meas. channels
 B^+ product
 B_2^+ product

Summary

- **Negative Ions are Strongly Correlated Systems and have Dramatic Relaxation Mechanisms; They present Serious Challenges to Theorists.**

- **Wigner Law is Valid at Threshold Despite PCI Effects as well as for Charge States**

- **Shape and Feshbach Resonances have been Observed in Light and Heavy Systems.**

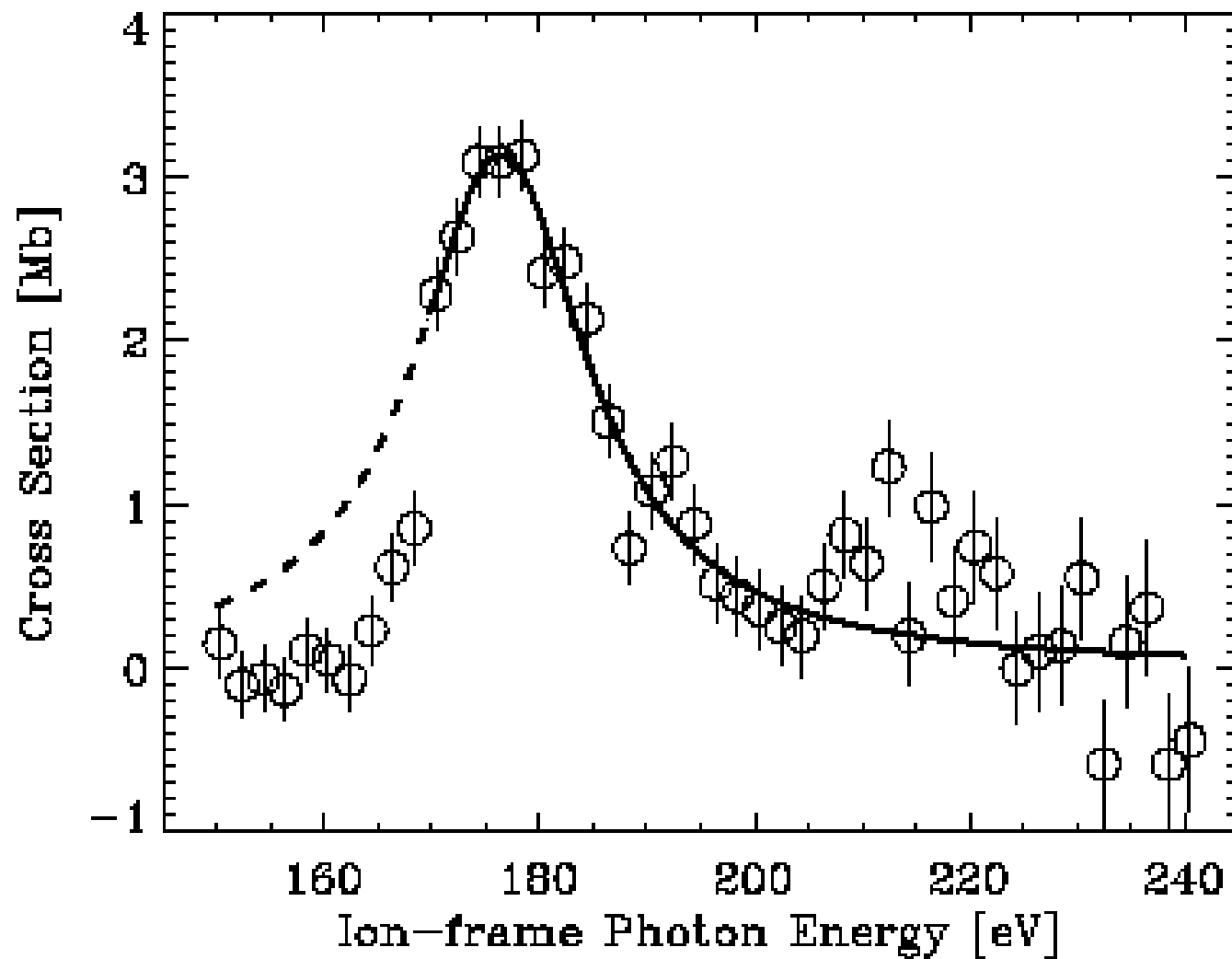- **Highly Charged Ions may be Produced via Shake-Off, Knock-out, Simultaneous**

2p Photodetachment Allows Charge State Formation

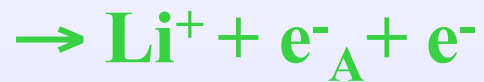Si^{4+}

Simultaenous Multi-Auger Decay???

Shape Resonance Observed Only in the S$^+$ Channel

K-Shell Photodetachment of Li⁻

$$h\nu + Li^- \rightarrow Li + e^-$$
$$\rightarrow Li^* + e^-$$
$$\rightarrow Li^+ + e^-_A + e^-$$

Probing Shape Resonance
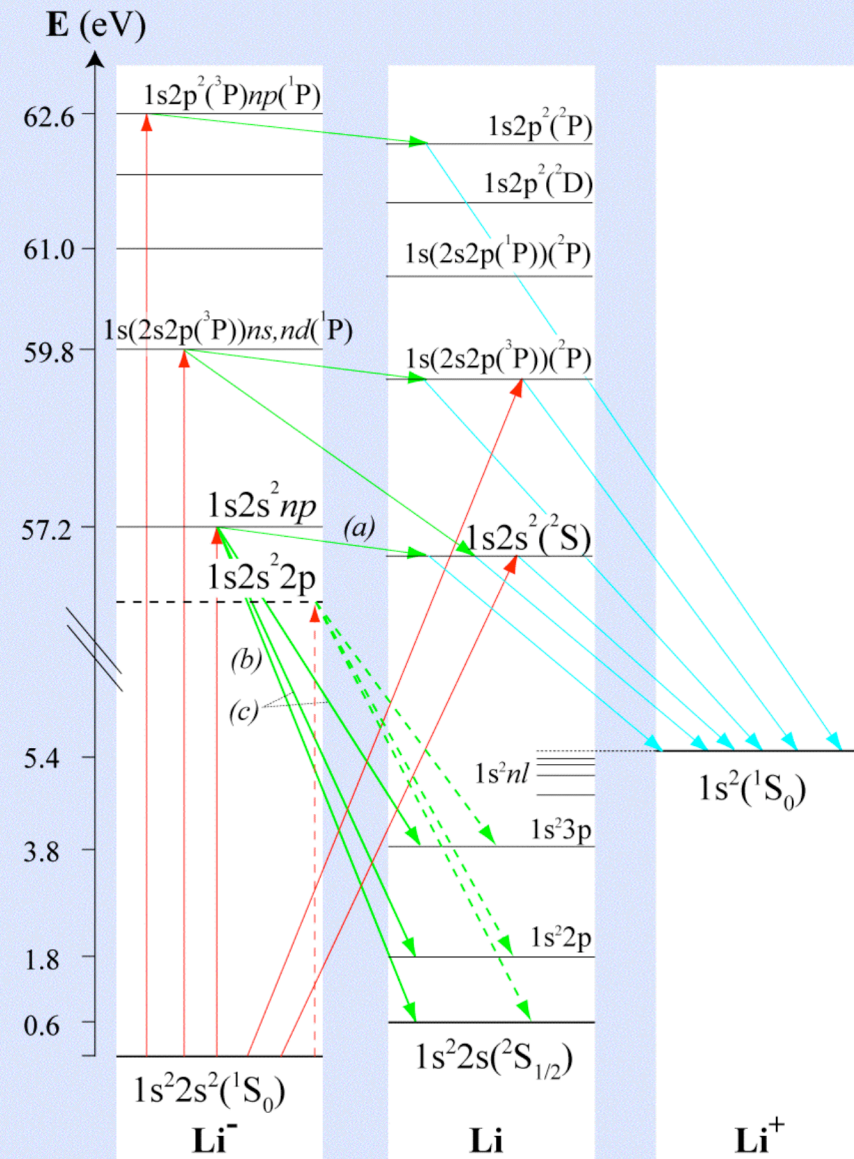
E (eV)

62.6	$1s2p^2(^3P)np(^1P)$
61.0	
59.8	$1s(2s2p(^3P))ns, nd(^1P)$
57.2	$1s2s^2 np$
	$1s2s^2 2p$
5.4	
3.8	
1.8	
0.6	

$1s^22s^2(^1S_0)$

Li⁻

$1s2p^2(^2P)$
$1s2p^2(^2D)$
$1s(2s2p(^1P))(^2P)$
$1s(2s2p(^3P))(^2P)$
$1s2s^2(^2S)$

(a)
(b)
(c)

$1s^2nl$
$1s^23p$
$1s^22p$
$1s^22s(^2S_{1/2})$

Li

$1s^2(^1S_0)$

Li⁺

Schematic energy level diagram. All excitations of Li⁻ have a propensity for decay into neutral Li and Li⁺. Only the notable decays are shown.

Recapture of the Photoelectron: Dramatic Post-Collision Recapture

Comparison between Experiment and Theories

Gorczyca et al.

— R-matrix
— R-matrix + classical damping

Cross Section (Mb)

Photon Energy (eV)

Berrah et al. PR. 87, 25002 (2001).

Gorczyca et al. PRA 68, 050703 (R), '03

Recapture of the Photoelectron: Dramatic Post-Collision Recapture (PCI)
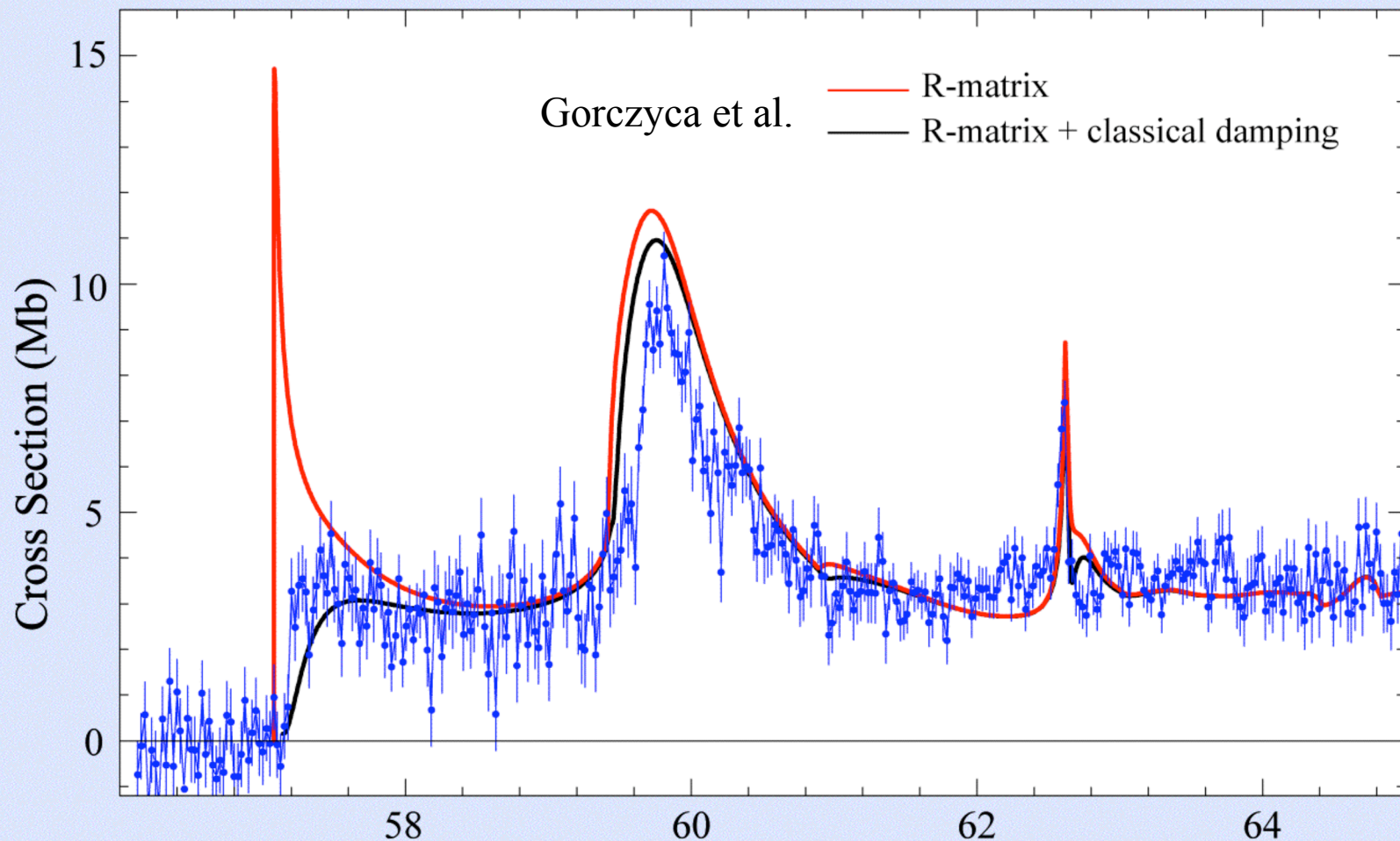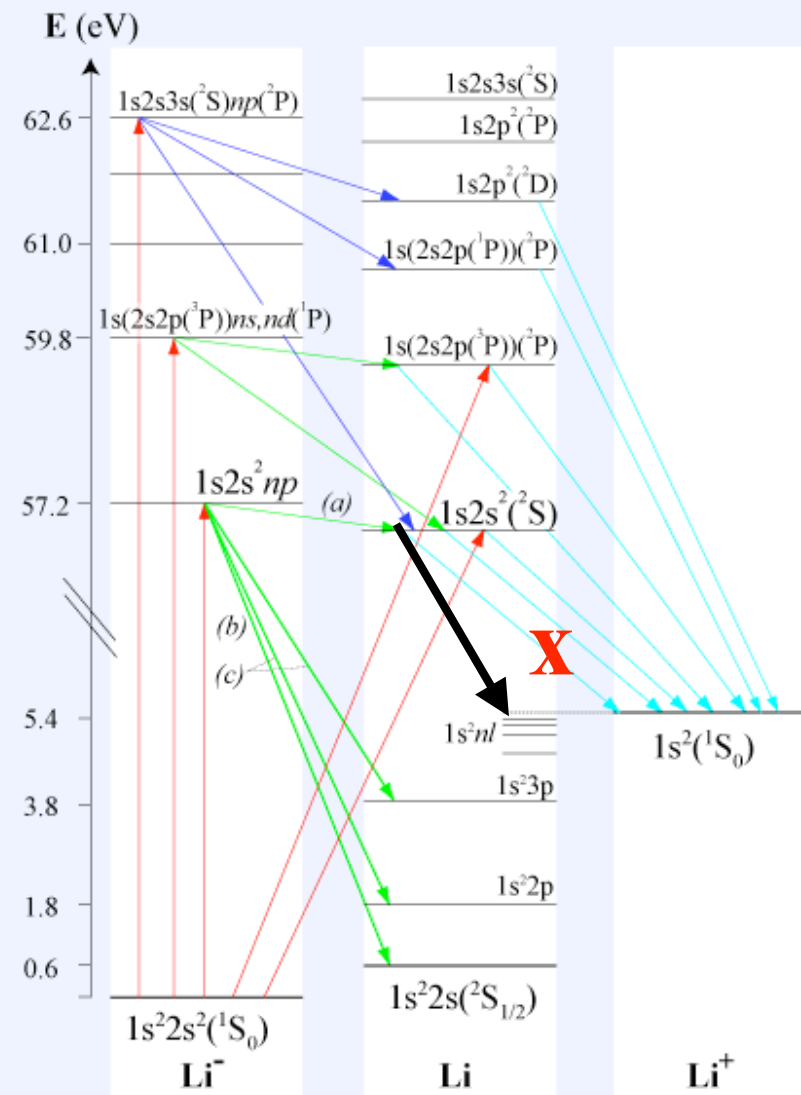

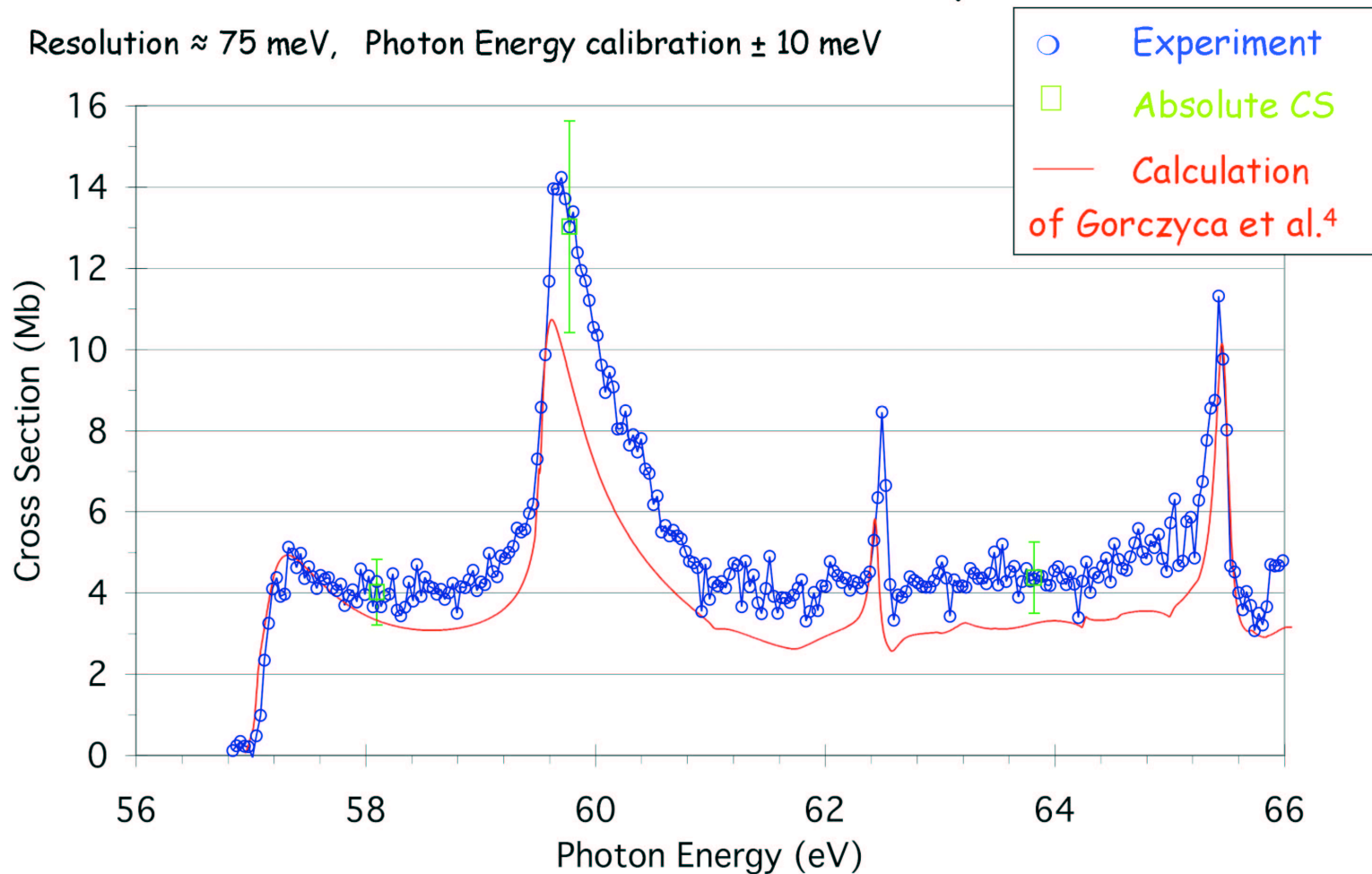Schematic energy level diagram. All excitations of Li⁻ have a propensity for decay into neutral Li and Li⁺. Only the notable decays are shown.

New Results: Absolute Cross section Measurements

Li⁻ Double-Photodetachment Spectrum

Resolution ≈ 75 meV,   Photon Energy calibration ± 10 meV

[4] T.W. Gorczyca, O. Zatsarinny, H.-L. Zhou, S.T. Manson, Z. Felfli, and A.Z. Msezane, *Phys. Rev. A* **68**, 050703 (2003)

Competition: Photodetachment and Photodissociation in B_2^-

$B_2^- + h\nu \rightarrow B_2^+ + 2e^-$

$B_2^- + h\nu \rightarrow B^+ + B + 2e^-$

$B_2^- + h\nu \rightarrow B^+ + B^- + e^-$

B^+ product
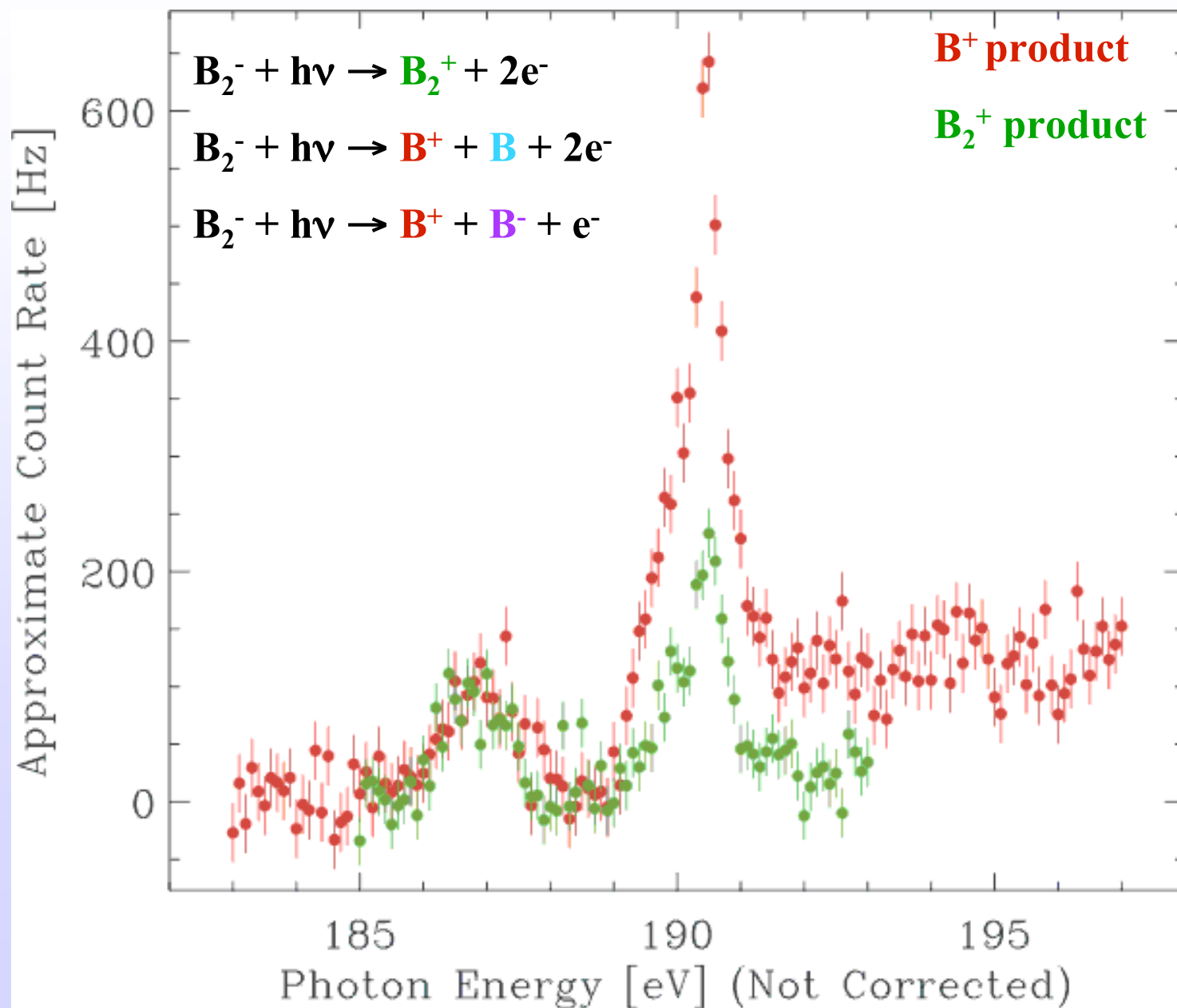
B_2^+ product

High Resolution Spectra for B_2^-

100 meV (nominal bandwidth)
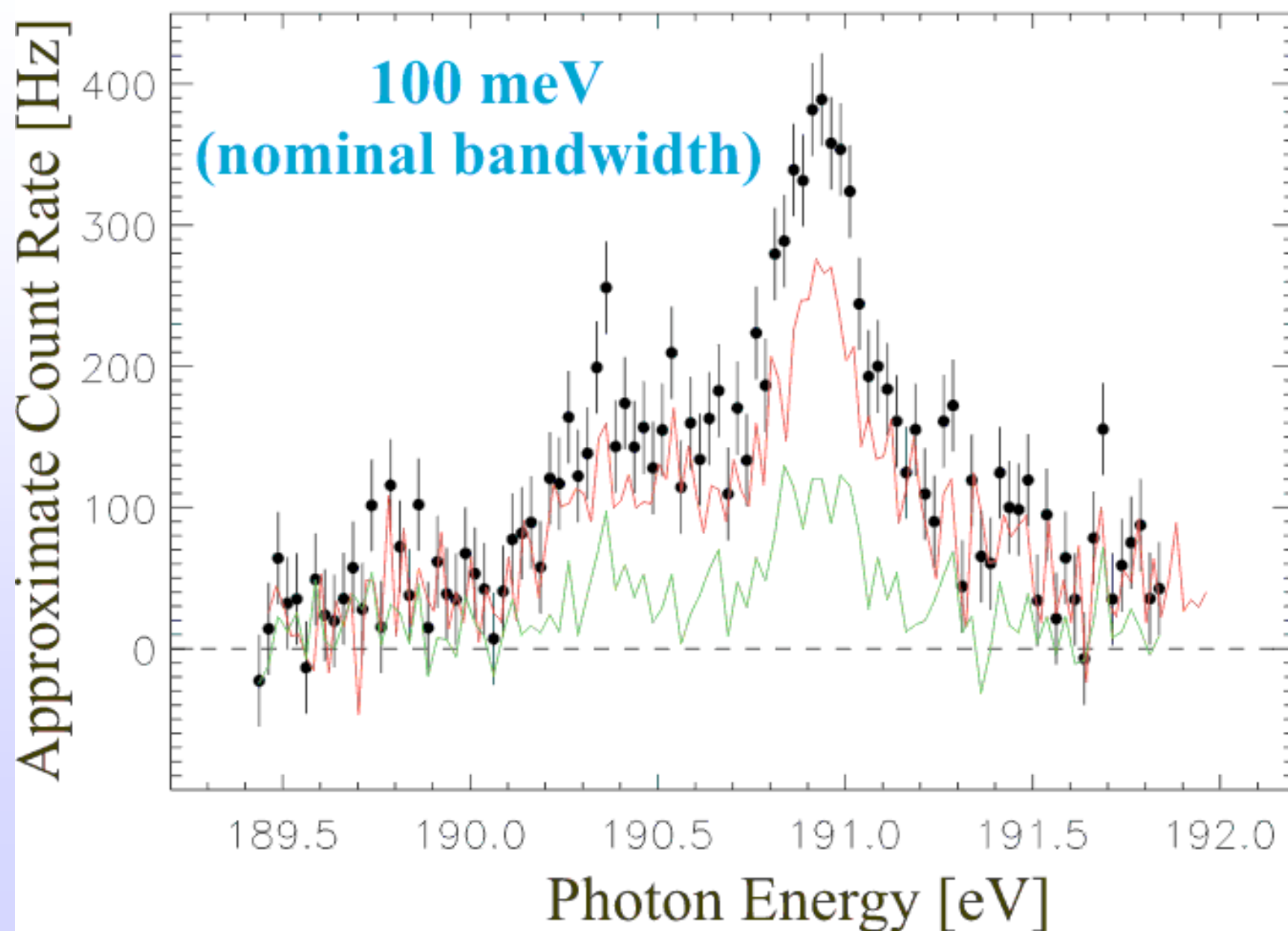
Photodetachment
&
Photodissociation
Reactions
Si⁻, Si₂⁻, Si₃⁻

Sum of channels
Si⁺ product
Si₂⁺ product
Si₃⁺ product
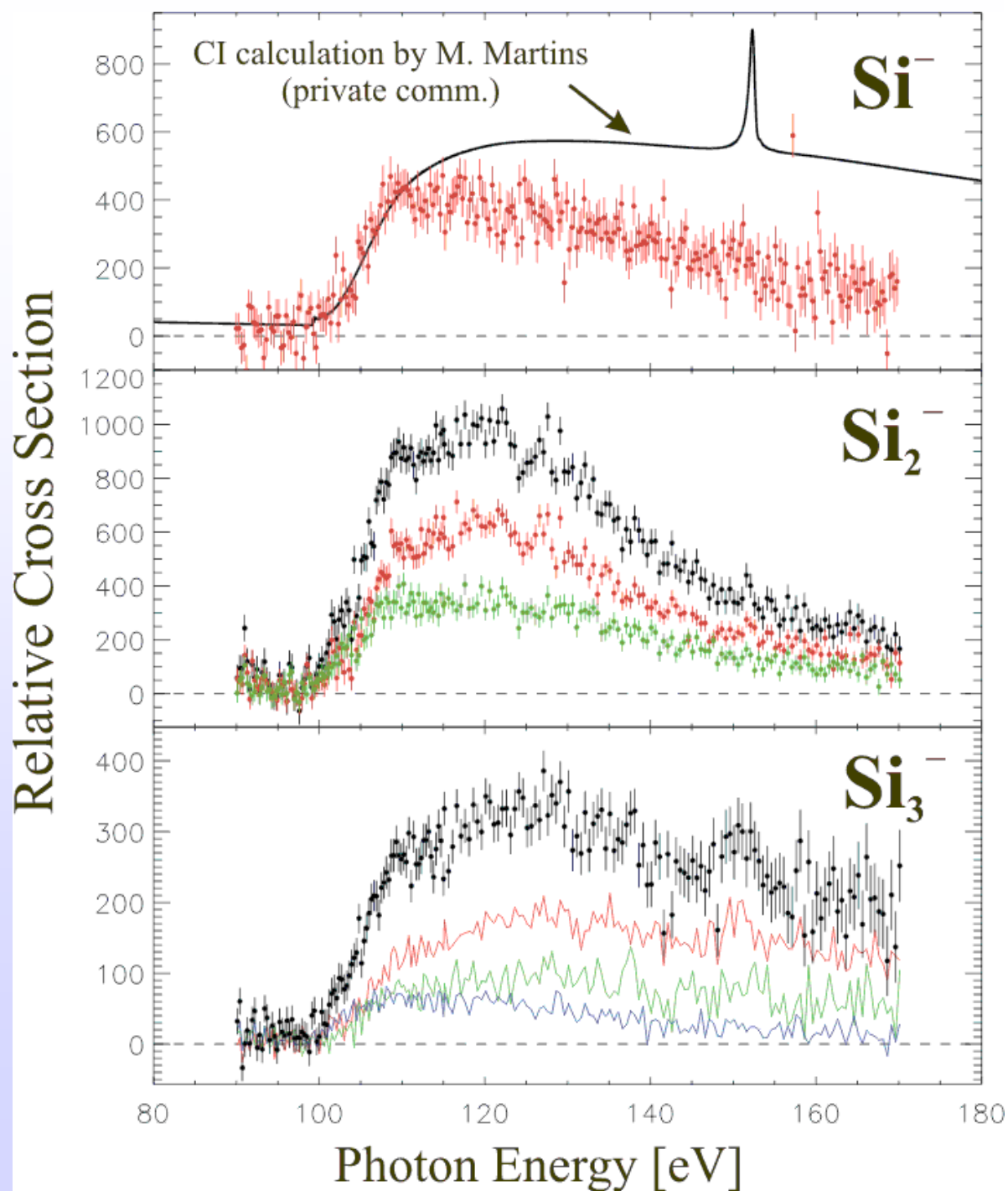
New Future AMO Scientific Directions: Complexity and Control

1. **Inner-Shell Studies of Metal Clusters (size dependent)**

2. **Two-Color Inner-Shell Experiments on Atoms, Molecules, Clusters and Ions (synch + ultrafast/high field lasers)**

3. **Study Atoms, Molecules, Ions and Clusters with the LCLS**

Highlights of New Future Research

1. Inner-Shell Studies of Cluster Anions:

- The Carbon Anion Chain...... C_{60}^{-}

- Transition Metals

  - Semi-conductor Ga_mX_n (X=P, As; n,m=1-3)

  - Metal hydride such as MH^-, MH_2^-, MH_3- and MH_4^- (M= Sc_ Cu).

- Development/Use of Photoelectron Spectrocopy on Ions Studies.

New Future Studies of Clusters

- **<u>Electronic Properties of Clusters</u>**:
  - Angle-resolved, inner-valence and inner-shell photoelectron spectroscopic studies of van der Waals clusters.

  - Studies of Metal clusters, Va, Ti, Fe and their oxides, as a function of cluster size.

- **<u>Magnetic Properties of Clusters</u>**:
  - Spin-resolved measurements; application to the magnetic recording industry.

TABLE I: Measured ratio of channel strengths $[\sigma(S^{n+})/\sigma(S^{m+}) \times 100\%]$, reported to 1 SD.

Photon Energy [eV]	Ratio of Channel Strengths [in %]		
	S^{2+}/S^+	S^{3+}/S^+	S^{3+}/S^{2+}
169.911	21.81(160)	1.819(139)	8.22(61)
199.866	58.0(41)	8.67(57)	14.95(98)
229.820	67.0(49)	16.28(104)	25.18(166)

TABLE II: Measured absolute cross sections, reported to 1 SD.

Photon Energy [eV]	Cross Section [Mb]			
	S^+	S^{2+}	S^{3+}	S^{4+}
170.147	3.30(39)	0.696(85)	0.0585(67)[a]	
200.172	4.18(55)	2.38(31)	0.357(45)[a]	
230.197	3.83(60)	2.31(34)	0.587(84)[a]	~0.023[b]

[a]The S^{3+} cross sections are obtained from the combination of S^+ and S^{2+} cross sections and the channel ratios of Table L.

[b]The S^{4+} signal was found to be about 100 times weaker (within a factor of ~3) than S^{2+} for energies between 230 to 240 eV.

Shape Resonance Observed in the S⁺ Channel